

ATTORNEY DOCKET NO.: 05015.0365U1


App. Serial No.: 09/662,965

CONCLUSION

In light of the above Amendment and Remarks, Applicants respectfully request that the rejections be withdrawn.

Payment in the amount of \$850.00 for the One Month Extension of Time and the RCE filing fee is to be charged to a credit card and such payment is authorized by the signed, enclosed document entitled: Credit Card Payment Form PTO-2038. No additional fee is believed due; however, the Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 14-0629.

Respectfully submitted,
NEEDLE & ROSENBERG, P.C.


Jacqueline M. Hutter
Registration No. 44,792

CERTIFICATE OF FACSIMILE TRANSMISSION UNDER 37 C.F.R. § 1.8

I hereby certify that this AMENDMENT is being sent via facsimile transmission addressed to (703) 308-2395, ATTN: EXAMINER P. SHORT, Mail Stop RCE, Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450, on the date shown below.


Jacqueline M. Hutter

Date

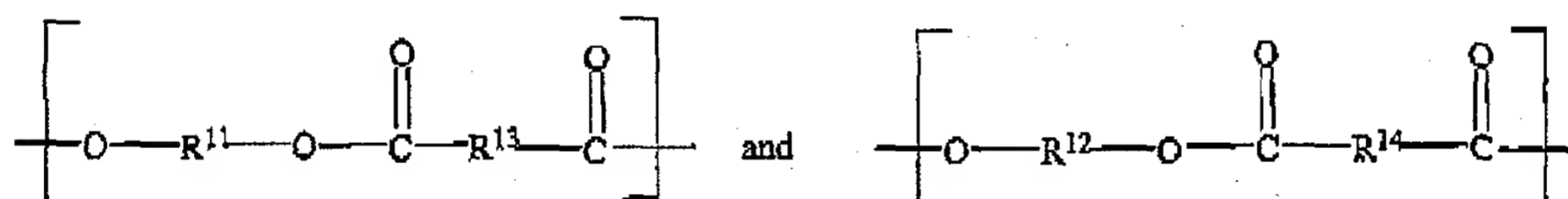
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ATTORNEY DOCKET NO.: 05015.0365U1

App. Serial No.: 09/662,965

MARKED-UP VERSION OF AMENDMENTS

1. A method for [slowing the degradation rate of] preparing a an article from a biodegradable polymer composition wherein the method comprises:
 - c. introducing a phenol-containing compound comprising terpene-phenol resin into a biodegradable polymer or biodegradable polymer composition in an amount sufficient to slow the degradation rate of the biodegradable polymer or biodegradable polymer composition; and
 - d. mixing the phenol-containing compound with the biodegradable polymer or biodegradable polymer composition;
 wherein the biodegradable polymer or biodegradable polymer composition comprises one or more of:
 1. an aliphatic-aromatic copolyester having repeat units of the following structures:



wherein

- (i) R^{11} and R^{12} are the same or different, and are residues of one or more of diethylene glycol, propylene glycol, 1,3-propanediol, 2,2-dimethyl-1,3-propanediol, 1,3-butanediol, 1,4-butanediol, 1,5-pentanediol, 1,6-hexanediol, 2,2,4-trimethyl-1,6-hexanediol, thiodiethanol, 1,3-cyclohexanedimethanol, 1,4-cyclohexanedimethanol, 2,2,4,4-tetramethyl-1,3-cyclobutanediol, triethylene glycol, or tetraethylene glycol;
- (ii) R^{11} and R^{12} are 100% of the diol components in the copolyester;
- (iii) R^{13} is absent or is selected from one or more of the groups consisting of $\text{C}_1 - \text{C}_{12}$ alkylene or oxyalkylene; $\text{C}_1 - \text{C}_{12}$ alkylene or

ATTORNEY DOCKET NO.: 05015.0365U1

App. Serial No.: 09/662,965

oxyalkylene substituted with one to four substituents

independently selected from the group consisting of halo, C₆ - C₁₀aryl, and C₁ - C₄ alkoxy; C₅ - C₁₀ cycloalkylene; and C₅ - C₁₀

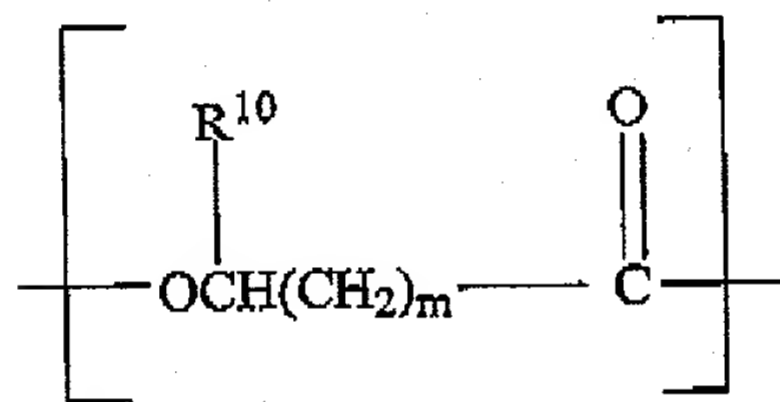
cycloalkylene substituted with one to four substituents

independently selected from the group consisting of halo, C₆ - C₁₀aryl, and C₁ - C₄ alkoxy; and(iv) R¹⁴ is selected from one or more of the groups consisting ofC₆ - C₁₀ aryl, and C₆ - C₁₀ aryl substituted with one to four

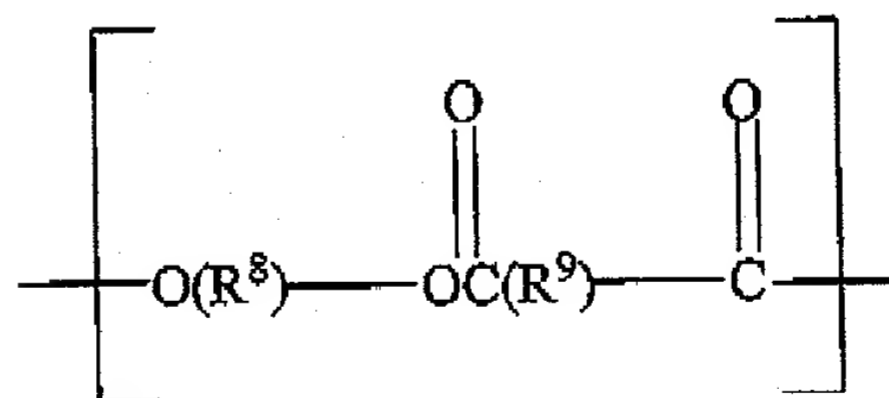
substituents independently selected from the group consisting of

halo, C₁ - C₄ alkyl, and C₁ - C₄ alkoxy;

2. an aliphatic polyester having repeat units of one or more of the following structures:



or



wherein m is an integer of from 0 to 10, and R¹⁰ is selected from the group consisting of hydrogen; C₁-C₁₂ alkyl; C₁-C₁₂ alkyl substituted with one to four substituents independently selected from the group consisting of halo, C₆-C₁₀ aryl, and C₁-C₄ alkoxy; C₅-C₁₀ cycloalkyl; and C₅-C₁₀ cycloalkyl

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NEEDLE & ROSENBERG

NO. 858

P. 26

EXHIBIT A

ATTORNEY DOCKET NO.: 05015.0365U1

App. Serial No.: 09/662,965

substituted with one to four substituents independently selected from the group consisting of halo, C₆-C₁₀ aryl, and C₁-C₄ alkoxy, wherein R⁸ is selected from the group consisting of C₂-C₁₂ alkylene or C₂-C₁₂ oxyalkylene; C₂-C₁₂ alkylene or C₂-C₁₂ oxyalkylene substituted with one to four substituents independently selected from the group consisting of halo, C₆-C₁₀ aryl, and C₁-C₄ alkoxy; C₅-C₁₀ cycloalkylene; C₅-C₁₀ cycloalkylene substituted with one to four substituents independently selected from the group consisting of halo, C₆-C₁₀ aryl, and C₁-C₄ alkoxy, and

wherein R⁹ is absent or is selected from one or more of the group consisting of C₁-C₁₂ alkylene or oxyalkylene; C₁-C₁₂ alkylene or oxyalkylene substituted with one to four substituents independently selected from the group consisting of halo, C₆-C₁₀ aryl, and C₁-C₄ alkoxy; C₅-C₁₀ cycloalkylene; and C₅-C₁₀ cycloalkylene substituted with one to four substituents independently selected from the group consisting of halo, C₆-C₁₀ aryl, and C₁-C₄ alkoxy; and

- 4) a C₁-C₁₀ cellulose ester having a DS equal to or less than about 2.5; and

c. forming the biodegradable polymer composition into an article, wherein the article comprises: a film, a bottle, a blow molded article, an injection molded article or a container.

7. (Twice Amended) The method of claim 1 wherein the biodegradable polymer or biodegradable polymer composition comprises the aliphatic-aromatic copolyester and wherein R¹¹ and R¹² are the same or different, and are selected from the group consisting of residues of one or more of diethylene glycol, propylene glycol, 1,3-propanediol, 1,3-butanediol, and 1,4-butanediol, R¹³ is selected from the group consisting of malonic acid, succinic acid, glutaric acid, adipic acid, pimelic acid, 2,2-dimethyl glutaric acid, diglycolic acid, and an ester forming derivative

ATTORNEY DOCKET NO.: 05015.0365U1

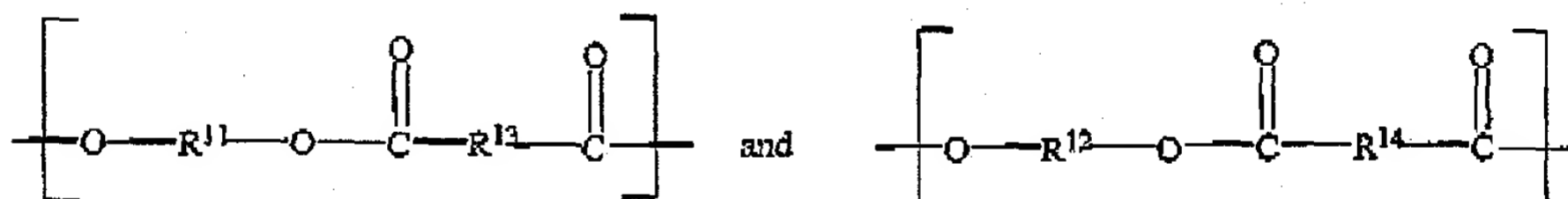
App. Serial No.: 09/662,965

thereof, and R^{14} is selected from the group consisting of one or more of 1,4-terephthalic acid, 1,3-terephthalic acid, 2,6-naphthoic acid, 1,5-naphthoic acid, and an ester forming derivative thereof.

10. (Twice Amended) A method for [slowing the degradation rate of] preparing an article from a biodegradable polymer or polymer composition, wherein the method comprises:

- (a) introducing a phenol-containing compound into a biodegradable polymer or polymer composition in an amount sufficient to slow the degradation rate of the biodegradable polymer or polymer composition; and
- (b) mixing the phenol-containing compound with the biodegradable polymer or polymer composition, wherein the biodegradable polymer comprises one or more of the following:

1. an aliphatic-aromatic copolyester having repeat units of the following structures:



wherein

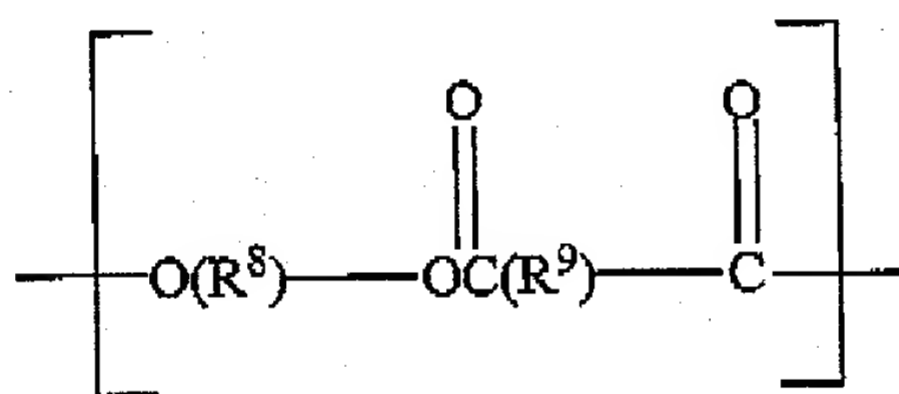
- (i) R^{11} and R^{12} are the same or different, and are residues of one or more of diethylene glycol, propylene glycol, 1,3-propanediol, 2,2-dimethyl-1,3-propanediol, 1,3-butanediol, 1,4-butanediol, 1,5-pentanediol, 1,6-hexanediol, 2,2,4-trimethyl-1,6-hexanediol, thiodiethanol, 1,3-cyclohexanedimethanol, 1,4-cyclohexanedimethanol, 2,2,4,4-tetramethyl-1,3-cyclobutanediol, triethylene glycol, or tetraethylene glycol;
- (ii) R^{11} and R^{12} are 100% of the diol components in the copolyester;
- (iii) R^{13} is absent or is selected from one or more of the groups consisting of $C_1 - C_{12}$ alkylene or oxyalkylene; $C_1 - C_{12}$ alkylene or

ATTORNEY DOCKET NO.: 05015.0365U1
App. Serial No.: 09/662,965

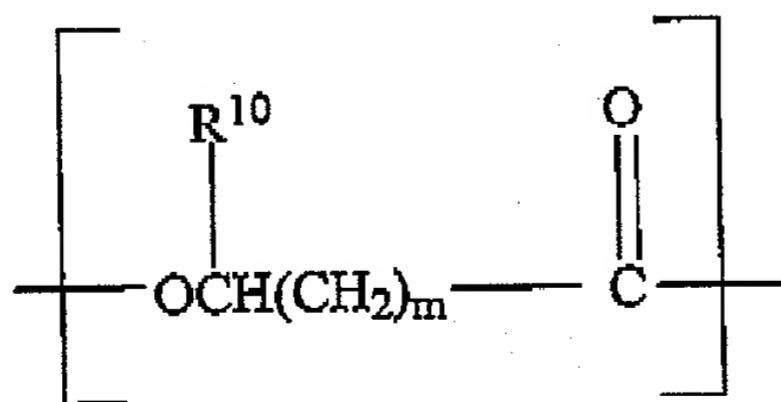
oxyalkylene substituted with one to four substituents independently selected from the group consisting of halo, C₆ - C₁₀ aryl, and C₁ - C₄ alkoxy; C₅ - C₁₀ cycloalkylene; and C₅ - C₁₀ cycloalkylene substituted with one to four substituents independently selected from the group consisting of halo, C₆ - C₁₀ aryl, and C₁ - C₄ alkoxy; and

(iv) R¹⁴ is selected from one or more of the groups consisting of C₆ - C₁₀ aryl, and C₆ - C₁₀ aryl substituted with one to four substituents independently selected from the group consisting of halo, C₁ - C₄ alkyl, and C₁ - C₄ alkoxy;

- 2) an aliphatic polyester having repeat units of one or more of the following structures:



or



ATTORNEY DOCKET NO.: 05015.0365U1

App. Serial No.: 09/662,965

wherein m is an integer of from 0 to 10, and R^{10} is selected from the group consisting of hydrogen; C_1 - C_{12} alkyl; C_1 - C_{12} alkyl substituted with one to four substituents independently selected from the group consisting of halo, C_6 - C_{10} aryl, and C_1 - C_4 alkoxy; C_5 - C_{10} cycloalkyl; and C_5 - C_{10} cycloalkyl substituted with one to four substituents independently selected from the group consisting of halo, C_6 - C_{10} aryl, and C_1 - C_4 alkoxy,

wherein R^8 is selected from the group consisting of C_2 - C_{12} alkylene or C_2 - C_{12} oxyalkylene; C_2 - C_{12} alkylene or C_2 - C_{12} oxyalkylene substituted with one to four substituents independently selected from the group consisting of halo, C_6 - C_{10} aryl, and C_1 - C_4 alkoxy; C_5 - C_{10} cycloalkylene; C_5 - C_{10} cycloalkylene substituted with one to four substituents independently selected from the group consisting of halo, C_6 - C_{10} aryl, and C_1 - C_4 alkoxy, and

wherein R^9 is absent or is selected from one or more of the group consisting of C_1 - C_{12} alkylene or oxyalkylene; C_1 - C_{12} alkylene or oxyalkylene substituted with one to four substituents independently selected from the group consisting of halo, C_6 - C_{10} aryl, and C_1 - C_4 alkoxy; C_5 - C_{10} cycloalkylene; and C_5 - C_{10} cycloalkylene substituted with one to four substituents independently selected from the group consisting of halo, C_6 - C_{10} aryl, and C_1 - C_4 alkoxy; and

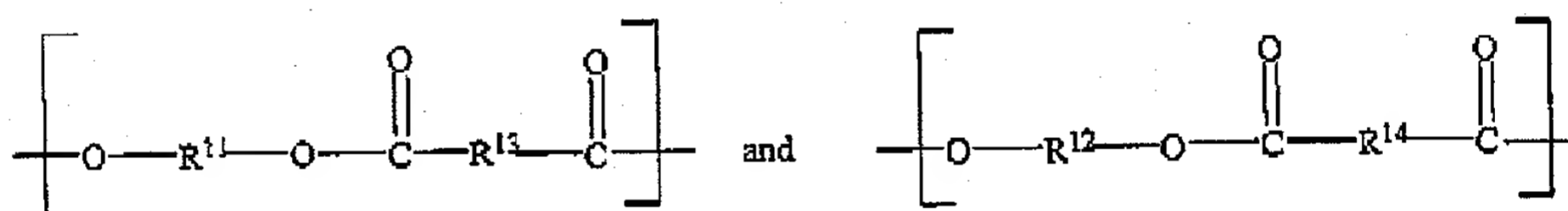
3) C_1 - C_{10} cellulose ester having a DS equal to or less than about 2.5; and

(c) forming the biodegradable polymer composition into an article, wherein the article comprises: a film, a bottle, a blow molded article, an injection molded article or a container.

23. (Twice Amended) A biodegradable polymer composition for making an article comprising a film, a bottle, a blow molded article, an injection molded article or a container, wherein the biodegradable polymer or biodegradable polymer-second material composition comprises [comprising]:

ATTORNEY DOCKET NO.: 05015.0365U1
App. Serial No.: 09/662,965

- a. a phenol-containing compound comprising terpene-phenol resin incorporated in the biodegradable polymer or biodegradable polymer-second material composition, the phenol-containing compound being present at an amount sufficient to slow the degradation rate of the biodegradable polymer or biodegradable polymer second-material composition; and
- b. a biodegradable polymer or biodegradable polymer-second material composition comprising one or more of the following:
 1. an aliphatic-aromatic copolyester having repeat units of the following structures:



wherein

- (i) R^{11} and R^{12} are the same or different, and are residues of one or more of diethylene glycol, propylene glycol, 1,3-propanediol, 2,2-dimethyl-1,3-propanediol, 1,3-butanediol, 1,4-butanediol, 1,5-pentanediol, 1,6-hexanediol, 2,2,4-trimethyl-1,6-hexanediol, thiodiethanol, 1,3-cyclohexanedimethanol, 1,4-cyclohexanedimethanol, 2,2,4,4-tetramethyl-1,3-cyclobutanediol, triethylene glycol, or tetraethylene glycol;
- (ii) R^{11} and R^{12} are 100% of the diol components in the copolyester;
- (iii) R^{13} is absent or is selected from one or more of the groups consisting of C_1 - C_{12} alkylene or oxyalkylene; C_1 - C_{12} alkylene or oxyalkylene substituted with one to four substituents independently selected from the group consisting of halo, C_6 - C_{10} aryl, and C_1 - C_4 alkoxy; C_5 - C_{10} cycloalkylene; and C_5 - C_{10} cycloalkylene substituted

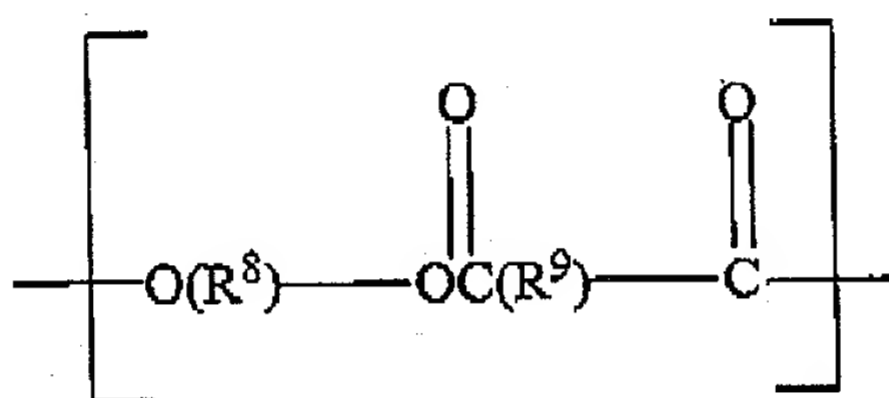
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App. Serial No.: 09/662,965

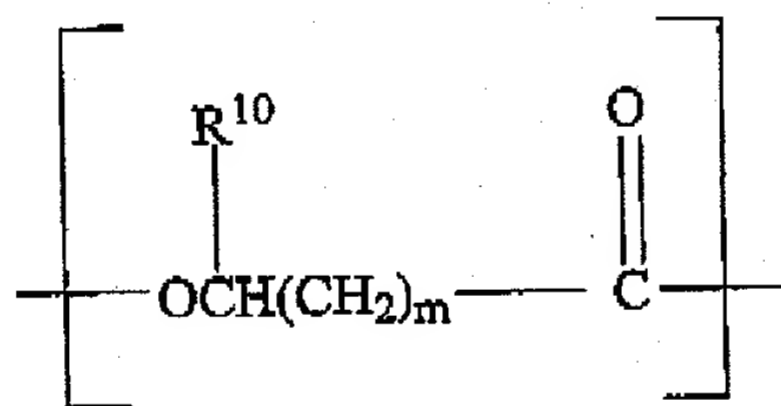
with one to four substituents independently selected from the group consisting of halo, C₆ - C₁₀ aryl, and C₁ - C₄ alkoxy; and

(iv) R¹⁴ is selected from one or more of the groups consisting of C₆ - C₁₀ aryl, and C₆ - C₁₀ aryl substituted with one to four substituents independently selected from the group consisting of halo, C₁ - C₄ alkyl, and C₁ - C₄ alkoxy;

- 2) an aliphatic polyester having repeat units of one or more of the following structures:



or



wherein m is an integer of from 0 to 10, and R¹⁰ is selected from the group consisting of hydrogen; C₁-C₁₂ alkyl; C₁-C₁₂ alkyl substituted with one to four substituents independently selected from the group consisting of halo, C₆-C₁₀ aryl, and C₁-C₄ alkoxy; C₅-C₁₀ cycloalkyl; and C₅-C₁₀ cycloalkyl substituted with one to four substituents independently selected from the group consisting of halo, C₆-C₁₀ aryl, and C₁-C₄ alkoxy,

ATTORNEY DOCKET NO.: 05015.0365U1

App. Serial No.: 09/662,965

wherein R^8 is selected from the group consisting of C_2 - C_{12} alkylene or C_2 - C_{12} oxyalkylene; C_2 - C_{12} alkylene or C_2 - C_{12} oxyalkylene substituted with one to four substituents independently selected from the group consisting of halo, C_6 - C_{10} aryl, and C_1 - C_4 alkoxy; C_5 - C_{10} cycloalkylene; C_5 - C_{10} cycloalkylene substituted with one to four substituents independently selected from the group consisting of halo, C_6 - C_{10} aryl, and C_1 - C_4 alkoxy, and

wherein R^9 is absent or is selected from one or more of the group consisting of C_1 - C_{12} alkylene or oxyalkylene; C_1 - C_{12} alkylene or oxyalkylene substituted with one to four substituents independently selected from the group consisting of halo, C_6 - C_{10} aryl, and C_1 - C_4 alkoxy; C_5 - C_{10} cycloalkylene; and C_5 - C_{10} cycloalkylene substituted with one to four substituents independently selected from the group consisting of halo, C_6 - C_{10} aryl, and C_1 - C_4 alkoxy; and

3) C_1 - C_{10} cellulose ester having a DS equal to or less than about 2.5.

24. The biodegradable polymer composition of claim 23 wherein the biodegradable polymer or biodegradable polymer-second material composition comprises the aliphatic-aromatic copolyester and wherein R^{11} and R^{12} are the same or different, and are selected from the group consisting of residues of one or more of diethylene glycol, propylene glycol, 1,3-propanediol, 1,3-butanediol, and 1,4-butanediol, R^{13} is selected from the group consisting of malonic acid, succinic acid, glutaric acid, adipic acid, pimelic acid, 2,2-dimethyl glutaric acid, diglycolic acid, and an ester forming

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VOLUME 23

SUGAR
TO
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nucleus using a ferrous compound, $\text{Na}_2\text{S}_2\text{P}_8\text{-Na}_2\text{SO}_3$ combination (197). In refining pulp from wood, the extent of pulp fiber damage can be diminished by pretreatment (198). Oxidative pretreatment of pine wood facilitates delignification (199). A process has been patented in which lignins are selectively isolated having molecular weights of over 5000 followed by reaction with Na_2SO_3 and an aldehyde (201). The derived lignosulfonate is useful as a dye dispersant. A method has been patented for producing amine salts of lignosulfonate (202). A pretreatment of cold soda pulp using HNO_3 and Cl_2 has been reported (203). A patent has been issued for the sulfation of lignin using organic amine SO_3 complex reagent (204). A process patent for sulfonating lignin using oleum below 40°C has been issued (205). In another study, SO_2 emissions were decreased by improving the combustion operation (207).

Other Lignin Sulfonation/Sulfation Developments. The use of lignin treatment procedures involving steam explosion techniques has received substantial attention (208-214). A process patent claims the use of sulfamic acid as an accelerating agent to separate lignin from cellulosic components (215). Another patent claims a process of alkoxylating alkali lignin followed by sulfation with SO_3 or ClSO_3H (216). Another discloses a process for producing oil-soluble sulfonated surfactants useful for oil recovery from lignin and alkylphenol (217). Most paper pulping processes generate SO_2 for use in sulfonation by burning molten sulfur.

Lignosulfonate Uses. Marketable lignosulfonates include ammonium, aluminum, calcium, chrome, ferrochrome, magnesium, potassium, sodium, and amine salts, and various combinations. Consumption of lignosulfonates may be divided into the following uses: animal feed pellets (15%), concrete additives (14%), road dust control (19%), oil-well drilling muds (4%), pesticide dispersant (8%), and other uses (45%) (218). Vanillin (3-methoxy-4-hydroxybenzaldehyde) is manufactured almost exclusively from lignosulfonate raw materials (219,220). The use of lignosulfonate was shown to be a sacrificial adsorbate for reducing petroleum sulfonate adsorption by 50% in enhanced oil recovery (221). However, it is effective only as a preflush, not as a cosurfactant in the sulfonate slug.

Styrene and Vinyl Monomer, Polymer, and Copolymer Sulfonates. The incorporation of sulfonates into polymeric material can occur either after polymerization or at the monomer stage (222). The sulfonic acid group is strongly acidic and can therefore be used to functionalize the polymer backbone to the desired degree. Depending on the molar fraction in the polymer, as well as on the macromolecular structure, the sulfonic acid group strongly interacts with water to bring about polymer swelling or gel formation, to a point where the complete dissolution of the polymer is possible (222). The ability of sulfonic acids to exchange counterions has made these polymers prominent in industrial water treatment (qv) applications, such as ion-exchange (qv) resins for demineralization, membranes for reverse osmosis or Donan dialysis, separators in electrochemical cells, and selective membranes of many types (222). Being strongly polar, the sulfonate functionality is used in such diverse areas as textile fiber dyeability, in thickeners and flocculants, rubber modifiers, and adhesive promoters (222). In addition, sulfonic acid derivatives, such as sulfonyl chlorides, amides, and anhydrides, make for an even wider range of uses.

The simplest monomer, ethylenesulfonic acid, is made by elimination from sodium hydroxyethyl sulfonate and polyphosphoric acid. Ethylenesulfonic acid

AUG. 12. 2003 11:56AM

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NO. 858

P. 30

EXHIBIT B

Ullmann's Encyclopedia of Industrial Chemistry

Sixth, Completely Revised Edition

Volume 36

Textile Auxiliaries
to
Tin, Tin Alloys, and
Tin Compounds

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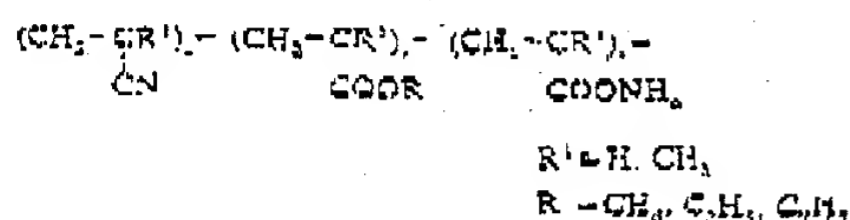
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Vol. 35

Textile Auxiliaries

25



On drying, ammonia is cleaved from the water-soluble ammonium polyacrylate, converting it to the water-insoluble, almost nonswelling acid form. A polyester or polyamide yarn sized in this way can be woven on water jet looms. The sizing agent can then be washed off under alkaline conditions.

Uses. Unlike other staple fiber sizing agents, which are usually supplied as the 100% solid product, poly(meth)acrylates are usually produced as a 25% aqueous solution, and have a wide range of uses owing to their wide variability. Like poly(vinyl alcohol), they are used as a high-quality additive in sizing formulations for all cotton yarns, especially for blends of cotton with synthetic fibers, and also for viscose staple fiber, wool, and polyacrylonitrile. Poly(acrylic acid) is the preferred sizing agent for polyamide filament yarns. Products based on acrylic acid copolymers are suitable for viscose, acetate, and triacetate filament yarns, while the hydrophobic poly(meth)acrylates based on (meth)acrylic esters are preferred for sizing polyester filament yarns. The excellent water-solubility of the sizing agents based on (meth)acrylic acid means that the gray cloth can be treated in subsequent processes without first desizing it. Recent research has shown that hydrophobic polyacrylate sizing agents are suitable for recovery and concentration by ultrafiltration [148].

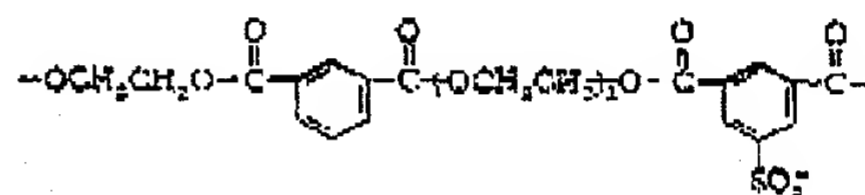
Ecology. Poly(meth)acrylates are practically biologically nondegradable. Hydrophobic methacrylic acid ester copolymers are adsorbed by the activated sludge, and can be eliminated in this way, while conventional hydrophilic products are eliminated to only a small extent [152, 154]. COD values of poly(meth)acrylate sizing agents are in the range ca. 1300–1700 mg O₂/g, based on solids material. Their aquatic toxicity is low, like that of the other sizing agents.

3.4.3. Polyester Condensates

With the introduction of the single-end sizing process [156], it became possible to apply water-

dispersible sizing agents based on polyesters. Today, these are the standard sizing agents for plain polyester filament yarns.

Polyester sizing agents are, in general, polycondensates of aromatic dicarboxylic acids with diols (e.g., ethylene glycol, diethylene glycol, or cyclohexane dimethanol) and sulfonated aromatic dicarboxylic acids (e.g., 5-Na-sulfoisophthalic acid), which provides solubility or dispersibility in water [157, 158]:



The proportion of ionic sodium sulfonate groups is such that the polyester condensates do not form true solutions in water, but dispersions of small particle size.

The properties of the polyester sizing agents can be controlled over a wide range by variation of the monomers, their relative proportions, and the molecular mass. The aim is to produce products that disperse readily in water and give films that are tough, resilient, and do not adhere to each other at high humidities. Polyester sizing agents have a chemical affinity to the polyester fiber and adhere to it strongly [126]. The possibility of interaction with the spinning preparation must be taken into account [159].

Uses. The main use of polyester sizing agents is for sizing plain polyester filament yarns. However, they are also used as a synergistic component to increase the adhesive strength when sizing staple fiber yarns made of polyester blends [160]. Note that when polyester sizing agents are washed out of the textile, there is a danger that they may be precipitated by electrolytes, especially polyvalent cations [161]. They can be determined on the textile by color reactions with basic dyes [134, 162, 163].

3.4.4. Other Synthetic Sizing Agents

Apart from the synthetic sizing agents described above, vinyl acetate copolymers have some practical importance. These consist of copolymers of vinyl acetate with acidic comonomers such as

AUG. 12. 2003 11:57AM

NEEDLE & ROSENBERG

NO. 858

P. 34

EXHIBIT C

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it is free from real or perceived adverse environmental effects, before global acceptance can become a reality. Thus, environmentally degradable polymers and plastics must meet very stringent guidelines for acceptance by a wide-ranging panel of reviewers. The importance of meeting this requirement is reflected in the search for acceptable definitions for environmentally degradable polymers, and new, more meaningful laboratory testing protocols for quantitatively measuring degradation and environmental fate and effects, and correlating the results of these experiments with real-world exposures. Consequently, definitions and test methods are addressed early in this article, prior to describing the important synthetic approaches under evaluation for environmentally degradable plastics and polymers and identifying some current and potential commercial products.

DEFINITIONS

There have been numerous definitions proposed for environmentally degradable plastics and polymers. ASTM D1566 defines a polymer as "a macromolecular material formed by the chemical combination of monomers having either the same or different chemical composition"; a plastic, as defined by ASTM D1695, is "a material that contains as an essential ingredient an organic substance of large molecular weight, is solid in its finished state, and, at some stage in its manufacture or in its processing into finished articles, can be shaped by flow." Definitions are important because they are indicative of expectations for the acceptance of environmentally degradable polymers and of the types of testing protocols that are needed to establish the acceptability of the polymers in the environment. The definitions developed by the American Society for Testing and Materials, ASTM D3883-93, for degradable, biodegradable, hydrolytically degradable, and oxidatively degradable plastics and given here are probably the most widely accepted, either as written or in some slightly modified form. They are equally applicable to polymers, in general.

Degradable plastic is a plastic designed to undergo a significant change in its chemical structure under specific environmental conditions, resulting in a loss of some properties that may vary as measured by standard test methods appropriate to the plastic and the application in a particular period of time that determines its classification.

Biodegradable plastic is a degradable plastic in which the degradation results from the action of naturally occurring microorganisms such as bacteria, fungi, and algae.

Hydrolytically degradable plastic is a degradable plastic in which the degradation results from hydrolysis.

Oxidatively degradable plastic is a degradable plastic in which the degradation results from oxidation.

Photodegradable plastic is a degradable plastic in which the degradation results from the action of natural daylight.

The definitions do not quantify the extent of degradation by any of the pathways and indicate only the mechanism that is operating to promote degradation. Although this is acceptable in a scientific sense in order to define the chemical process, it does not really go far enough to satisfy the requirements for environmentally acceptable polymers (19), which in the minds of legislators and

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P. 38

EXHIBIT D